Experimental report

08/02/2018

Proposal: **5-41-911**

Council: 10/2016

Title: Dimerization in the commensurate antiferromagnetic phase of NaFe(WO4)2

Research area: Physics

This proposal is a new proposal

Main proposer: **Sebastian BIESENKAMP**

Experimental team: Sebastian BIESENKAMP

Local contacts: Navid QURESHI

Samples: NaFe(WO4)2

Instrument	Requested days	Allocated days	From	To
D10	6	5	17/02/2017	22/02/2017

Abstract:

NaFe(WO4)2 is closely related to MnWO4 one of the prototype spiral multiferroics, and it exhibits transitions from incommensurate cycloid order to a commensurate up-up-down-down structure, which appears in multiferroic MnWO4 and RMnO3. Thermal expansion measurements reveal very strong anomalies upon entering the commensurate phases, which together with the magnetic frustration suggests that structural dimerization accompanies the antiferroamagnetic order. We propose to determine this structural dimerization in the commensurate magnetic phase.

Experimental report 5-41-911

The main goal of this experiment was to find an experimental proof of a dimerization effect in $NaFe(WO_4)_2$ and to investigate its strength in the different reported magnetic phases. A comprehensive study of the magnetic phases can be found in [1]. For increasing anharmonic contributions in the incommensurate order and for transitions into the commensurate phases (HF-C and LF-C), $NaFe(WO_4)_2$ very strong thermal expansion anomalies, as the b parameter shrinks by $\Delta b/b \approx 2.6 \times 10^{-4}$. The magnetic up-up-down-down structure with alternating ferromagnetic and antiferromagnetic arrangements is expected to cause different bond angles between Fe^{3+} and O^{2-} ions. These alternating bond angles induce a dimerization, which can be compared to the dimerization of a spin-Peierls transition for half integer spins and the associated bond-angle variation in $CuGeO_3$ [2]. A magnetoelastic modulation of the magnetic interaction would induce a structural distortion, which itself generates structural superstructure reflections. Such structural distortion should be related to the observed large thermal expansion anomalies. As the magnetic Fe^{3+} ions are building zig-zag chains along the c direction, it is expected to loose the c-glide plane in the case of a dimerization effect. This entails occurring superstructure reflections at $\mathbf{Q} = (h\,0\,l = \text{odd})$ and hence, an experimental proof of this dimerization effect can be carried out by measuring the occurrence of those reflections in the different magnetic phases of $NaFe(WO_4)_2$.

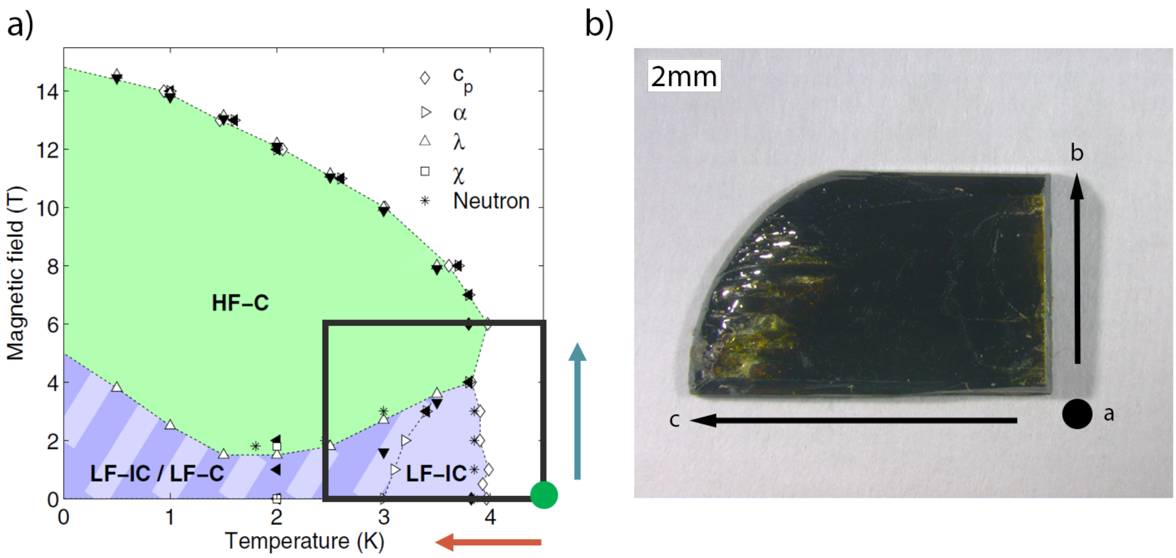

Figure 1.1: Phase diagram and sample of $NaFe(WO_4)_2$
Figure a) displays the magnetic phase diagram of $NaFe(WO_4)_2$ (adapted from [1]). Red and blue arrows mark clockwise and counterclockwise temperature and magnetic field dependent sweeps through the magnetic phase diagram. Both sequences start from the green dot. Figure b) displays the measured sample.

In the first part of the allocated beamtime, rocking scans over allowed reflections and possible superstructure reflections have been carried out in the paramagnetic phase. Even in the paramagnetic phase, reflections at $\mathbf{Q} = (h0l = \text{odd})$ can be detected. These normally forbidden reflections in the paramagnetic phase can be attributed mainly to multiple reflection. Thus, an occurring superstructure reflection resulting from a dimerization effect would add intensity on top of the multiple reflection peak. Hence, the intensity difference between the magnetic and paramagnetic phase for corresponding reflections is the crucial quantity to confirm the dimerization effect. These first rocking scans have been executed by using the area detector. As a weak signal and a large background prevent the observation of an intensity difference for $\mathbf{Q} = (h\,0\,l = \text{odd})$ reflections, the area detector has been replaced by a single ^{3}He detector and an additional analyzer has been mounted. Two of the measured reflections with the adapted instrumental setup are plotted in figure 1.1. With respect to the paramagnetic phase, an intensity enhancement can be observed for the measured reflections inside the HF-C phase and this intensity difference corresponds to the intensity of a superstructure reflection.

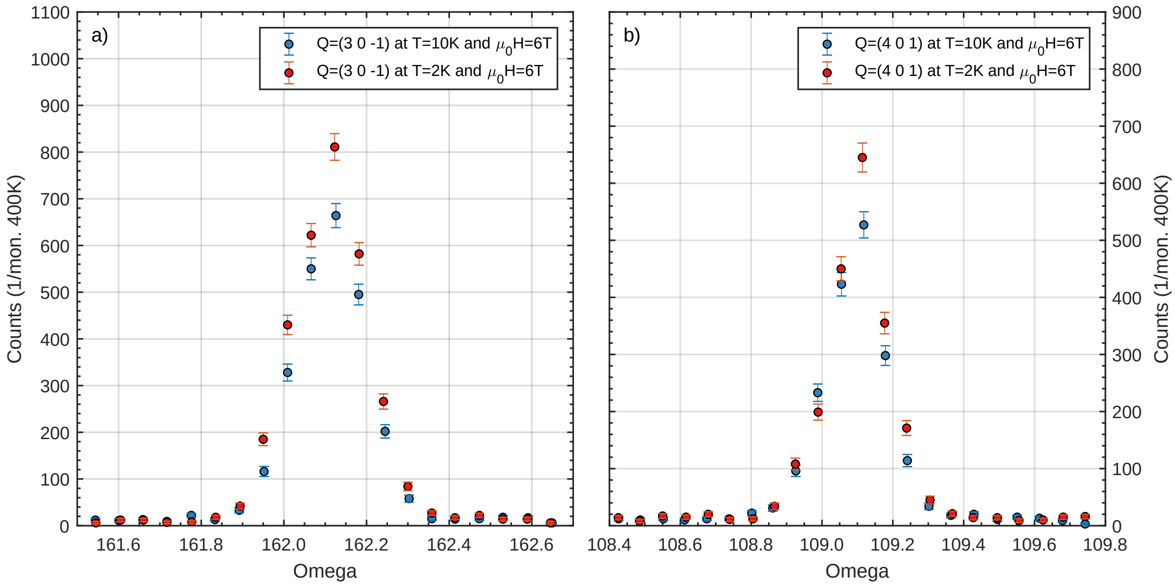

Figure 1.2: Measurements with an analyzer and a single ^{3}He detector
Both figures show measurements of $\mathbf{Q} = (h\,0\,l = \text{odd})$ reflections with higher statistic and lower background, as the counting time had been enhanced and an additional analyzer had been deployed. Referring ω scans have been executed in and outside the commensurate phase.

The intensity difference that is supposed to come from an evolving superstructure reflection has been determined to be rather small compared to typical Bragg reflections. Temperature and magnetic field dependent sweeps through the phase diagram have been executed in order to document the strength of the dimerization effect in the different magnetic phases. Clockwise and counterclockwise sweeps through the phase diagram have been started from the green marked point in the phase diagram in figure 1.1 and are marked in red and blue respectively. Both kinds of sweeps have been executed, because the accessibility of the LF-C and LF-IC phase depends on the sweep direction [1]. The results are plotted in figure 1.3 exemplary for the $\mathbf{Q} = (3\,0\,-1)$ reflection. From the recorded data, it can bee seen that a superstructure intensity emerges in the HF-C and LF-C phases and that the effect is strongest for the LF-C phase, where system is in its ground state [1].

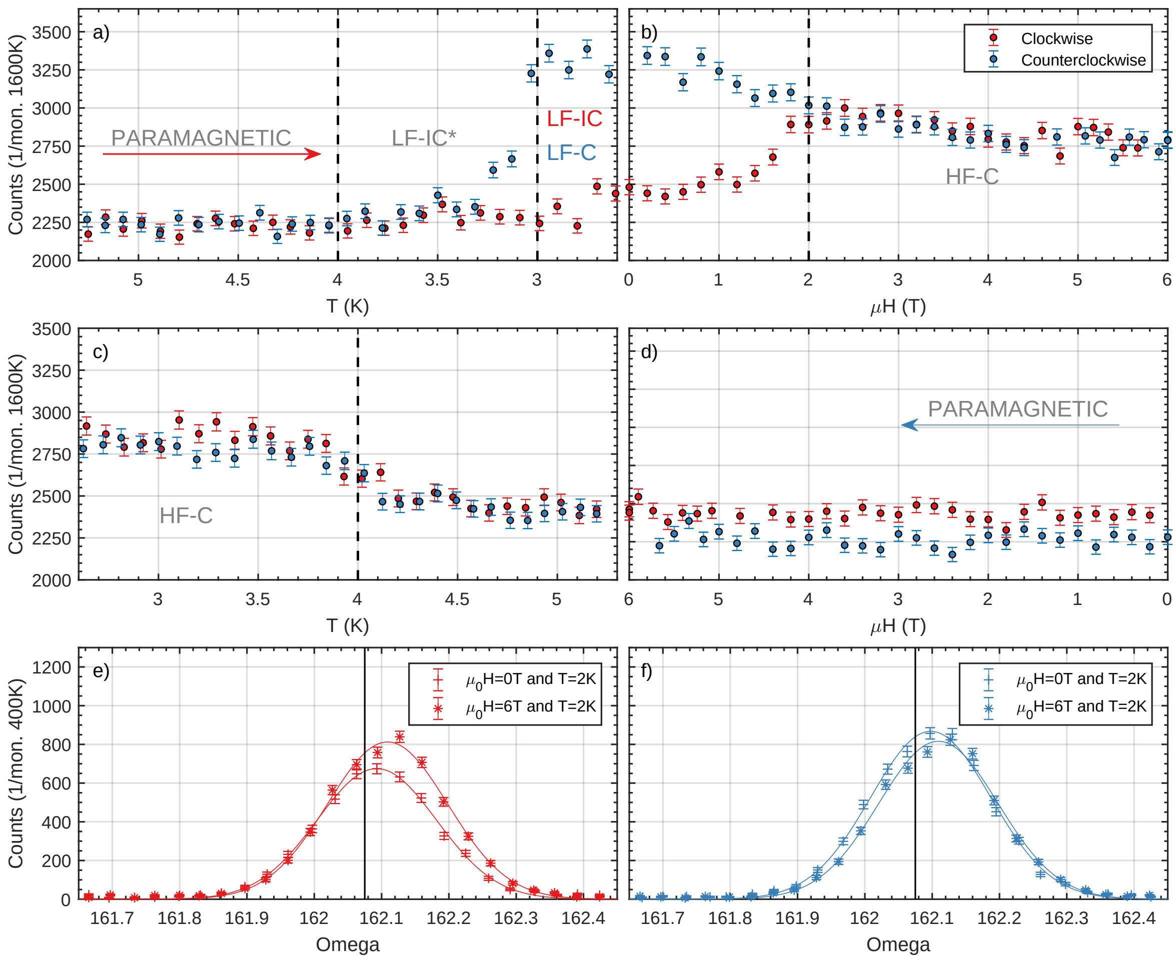

Figure 1.3: Temperature and magnetic field dependent sweeps for $\mathbf{Q} = (3\,0\,-1)$ Figure a)-d) display the temperature and magnetic field dependent sweeps trough the phase diagram for $\mathbf{Q} = (3\;0\;-1)$. The plots in e) and f) show the peak-center shift for different positions in the magnetic phase diagram. Figure e) corresponds to the clockwise motion and f) to the counterclockwise motion. The black line corresponds to the fixed ω value during the counting time. A peak center shift is present but not responsible for the observed enhancement of intensity.

During the experimental course of this beamtime, it was possible to record an experimental proof of a dimerization effect in the commensurate phases of $NaFe(WO_4)_2$. Due to finite beamtime, it was not possible to measure as many reflections, as it is necessary for a structural refinement. In order to get quantitatively information about the dimerization and the structural distortion, a continuation of this beamtime would be highly beneficial and would complete the investigations of the dimerization effect in $NaFe(WO_4)_2$.

References

[1] S. Holbein, et al. *Phys. Rev. B*, 94:104423, 2016.
[2] M. Braden, et al. *Phys. Rev. B*, 54(2):1105–1116, 1996.